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## ATMOSPHERIC DIFFUSION OF DROPLET CLOUDS

Jerome Aroesty

July 1984

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Prepared for

The Defense Advanced Research Projects Agency

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A preliminary analysis of contamination footprints associated with release of liquid chemical warfare agents in the atmosphere. When defense systems analysts require contamination footprints for situations that depart radically from the limited set of available field data, they use computer models presumably based on a sound understanding of the underlying physics to predict contamination zones and magnitudes. The author has found, however, that the dispersal and diffusion models most used in air base and tactical ballistic missile defense studies neglect a fundamental element in describing the processes of atmospheric diffusion. This omission translates into a potentially severe underestimate of toxic cloud exposure. It is shown how these models can be modified to include the important effect of the droplets' fall velocity.

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## PREFACE

An accurate representation of the physics of atmospheric diffusion is required to narrow the uncertainties found in models of air base defense against chemical agents delivered by tactical ballistic missiles. This Note presents a discussion of the diffusion and dispersion of toxic droplets when these processes are affected both by atmospheric turbulence and by the droplets' own considerable fall velocity. It is shown how currently used models, which may neglect the influence of fall velocity, can easily be modified to include this important parameter.

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## SUMMARY

This Note summarizes a preliminary analysis of contamination footprints associated with release of liquid chemical warfare agents (C.W.) in the atmosphere. The study was suggested because defense systems analysts often require contamination footprints for situations that depart radically from the limited set of field data. Computer models, presumably based on a sound understanding of the underlying physics, are then used to predict contamination zones and magnitudes.

We found that the C.W. dispersal and diffusion models, developed by the Chemical Systems Laboratory (CSL) of the U.S. Army Armament Research and Development Command and used in air base and tactical ballistic missile defense studies by Rand, Institute of Defense Analyses, Hughes, General Research Corporation, Aerospace Medical Research Laboratory, and others, neglect a fundamental element in describing the processes of atmospheric diffusion. This omission translates into a potentially severe underestimate of C.W. deposition densities and a physically implausible picture of "toxic cloud" exposure. Further implications and extensions of our preliminary findings remain to be explored.

One explanation for the utility of the CSL computer models to defense analysts is the possibility of compensating errors--that the order of magnitude error in modelling diffusion is compensated by large omissions or cancelling errors in the other components of the models in order to fit a limited data set. If this is the case, physical understanding is inadequate but the results may be satisfactory for situations that are close to those of the few field tests. Otherwise, the models fail the test of theoretical plausibility by neglecting the influence of fall velocity on diffusion. It is hoped that a program of theory and experiment can narrow the range of uncertainty.

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## I. INTRODUCTION

Defense analysts are now interested in understanding the implications of a potential tactical missile threat that includes warheads containing chemical warfare (C.W.) agents intended for atmospheric dispersal. Quantitative understanding of this threat, for purposes of analysis and studying defense system effectiveness, relies heavily on our ability to predict the contamination footprint associated with the dispersal of such agents. Several factors that influence the biological effects of these agents help to define a contamination footprint. These factors include:

- Meteorological conditions.
- Physical, chemical, and rheological properties of the agents.
- Nature of the agent release process and the formation of droplets by aerodynamic stripping as influenced by dispense altitude and velocity.
- Formation of vapor clouds, and the tradeoff between liquid deposition and evaporation.
- Atmospheric dispersal and diffusion of droplets and vapor.

Because field data and observations are sparse, computer models of the underlying physical processes must be used to develop a picture of the dispersion, diffusion, and surface evaporation phenomena that influence the contamination footprint.

Accurate models of the atmospheric diffusion of toxic clouds of liquid C.W. droplets should be key ingredients in programs to compute contamination patterns and C.W. deposition rates. Such models have been developed by the British [1] over the past twenty-five years in the context of research on chemical defense.<sup>1</sup> Unfortunately, the widely used U.S. model NUSSE2 [2], developed by the Chemical Systems Laboratory (CSL) of the U.S. Army Armament Research and Development Command fails

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<sup>1</sup>See the appendix for a review of the limited theoretical and empirical data base.

to incorporate the British results in its current version, and as a consequence overestimates atmospheric diffusion. This overestimate yields C.W. deposition rates and contamination footprints that could be in error by an order of magnitude or more. As discussed below, we have made a preliminary study of the potential discrepancy. Figure 1 exhibits the magnitude of the possible error in terms of the deposition of an agent released from a point source at a height  $H$  above the ground. The lower curve, A, is a nondimensional representation of the deposition rate based on an atmospheric diffusion theory that is similar to that used in NUSSE2 and other CSL programs. The upper curve, B, is based on an atmospheric diffusion theory that incorporates a more plausible representation of the physics of atmospheric spreading of clusters of liquid droplets. There is a large difference in the deposition pattern: the theoretically correct pattern involves much higher rates of deposition (over smaller crosswind distances). The difference between the two curves stems from the following:

Curve A, the lower one, assumes that liquid droplets, as they fall through the atmosphere, randomly diffuse like parcels of air about their centroid--their diffusion characteristics are assumed to be the same as tracers with air-like mass, and are entirely influenced by the turbulent eddy motion of air parcels.

Curve B, the upper one, reflects the finite mass of the falling droplets to the extent that their considerable terminal fall velocity inhibits the random diffusive motion about their centroid. Thus C.W. droplets, as they fall, are only slightly buffeted by the air's eddy motion and in fact tend to fall through or cross the trajectory of eddies rather than follow them.

C.W. droplets that are large enough to evaporate slowly and to survive until ground impact tend to be between one-half millimeter and five millimeters in diameter, and have terminal vertical or fall velocities between 1 and 10 m/sec. For these droplets, the true diffusion process is up to an order of magnitude weaker than one computed by neglecting the fall velocity effect.

It is possible that the CSL models employ diffusion parameters that are thus unrealistically high in order to account for droplet depositions actually observed during the limited field tests. This

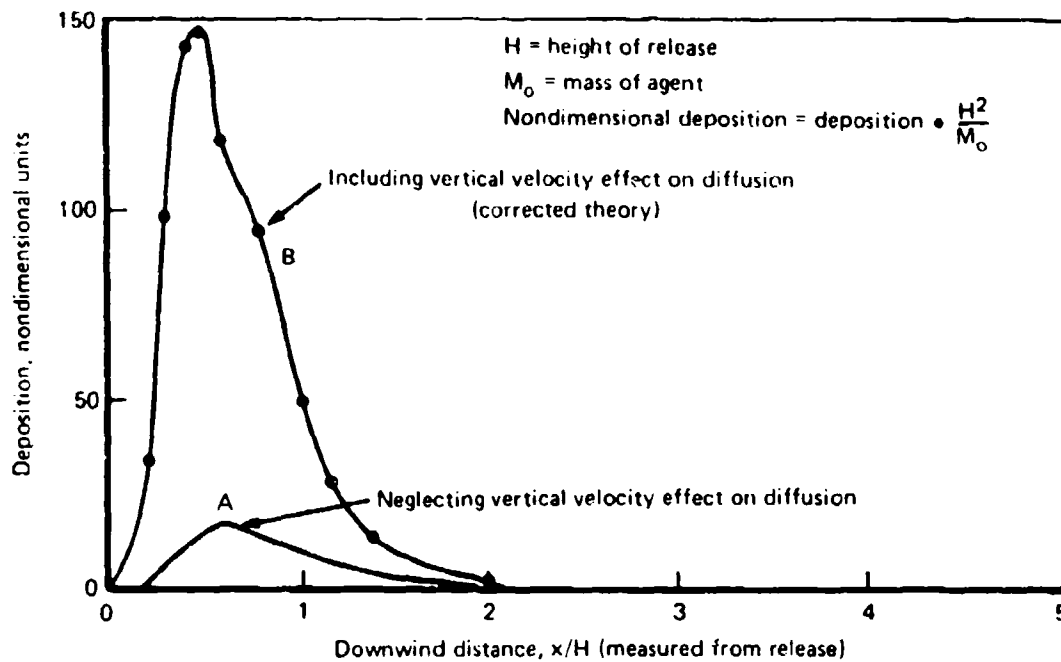


Fig. 1 — Deposition along centerline

implies that other phenomena, either omitted or not accurately described by the CSL models, were present during the field tests, and computer models rely on the "theory of compensating errors." Particle inertia, wind shear and the turning of the wind vector with altitude (Ekman spiral) are examples of phenomena which are known to influence diffusion, and which are not accounted for in the CSL model. However, none of these mechanisms are as important as the vertical velocity effect over the time scales associated with hypothetical C.W. release altitudes. Furthermore, all diffusion processes are expected to be strongly damped by the fall velocity effect.

## II. DISPERSION AND DIFFUSION

Atmospheric winds and turbulence cause clouds of liquid droplets to disperse and diffuse in two different ways.

1. Because the clouds include droplets of varying size and fall velocity, they tend to disperse ballistically in the downwind direction as drops fall and are carried by the wind. Small droplets travel further downwind than do larger droplets. This can be computed from droplet size distribution, wind speed, and direction. This is a deterministic process that we designate as ballistic dispersion, to distinguish it from diffusion.
2. The crosswind spreading is entirely due to the diffusion effect--even if all droplets were a single size they would still exhibit random motion and diffuse crosswind due to atmospheric turbulence. They would also diffuse isotropically about their ballistic trajectories. This can be accounted for by considering a polydisperse cloud of particles consisting of subclouds of uniform particles, each with a distinct fall velocity. The entire cloud is then accounted for by summing over the contribution due to the individual subclouds. Thus the diffusion process is linked to the dispersion process, and both must be described accurately.

If we consider a subcloud (or puff) of monodisperse droplets released at time  $t = 0$ , the concentration of agent is represented by a Gaussian spatial distribution with a single standard deviation  $\sigma(t)$  in all three directions, assuming isotropic turbulence and that the "falling droplet" effect is felt in  $x, y, z$  directions, or

$$\frac{N}{\sigma^3(2\pi)^{3/2}} \cdot \exp \left\{ - \left[ \frac{(x - x_0(t))^2 + (z - z_0(t))^2 + y^2}{2\sigma^2} \right] \right\} \quad (1)$$

where  $N$  is mass released,  $x$  is distance downwind measured from release,  $z$  is vertical distance,  $y$  is crosswind distance, and  $x_0(t)$ ,  $z_0(t)$ ,  $0$  are the coordinates of the centroid of the cloud. Note also that

$$\begin{aligned} x_0(t) &= \int_0^t u dt \text{ where } u \text{ is the horizontal windspeed,} \\ z_0(t) &= H - \int_0^t v dt \text{ where } v \text{ is fall velocity,} \\ &\text{and } H \text{ is the height of the release.} \end{aligned}$$

Smith and Hay [3] developed a theory that accounts for the dependence of  $\sigma$  on finite fall velocity. (See appendix for further discussion.) The theory is surprisingly tractable, but does not yield a simple explicit formula for  $\sigma(t)$ . Thomas [4] suggested that the Smith-Hay theory for diffusing falling particles can be simplified to derive the simple approximation (for preliminary applications and studies):

$$\sigma = \frac{(2/3) \cdot i^2 \cdot u^2 \cdot t}{\sqrt{v^2 + u^2/\beta^2}} \quad (2)$$

where  $\sigma$  is the standard deviation of the cloud

- $i$  is the turbulence intensity (magnitude about .1)
- $t$  is the time since release
- $v$  is the fall velocity
- $u$  is the wind speed
- $\beta$  is the ratio of Lagrangian and Eulerian scales (magnitude about 5).

If we designate  $\sigma^{\text{LIGHT}}$  as the value for light particles (which have negligible terminal velocity), then

$$\sigma/\sigma^{\text{LIGHT}} = \left(1 + \frac{v^2 \beta^2}{u^2}\right)^{-1/2} \quad (3)$$

Thus, if  $\beta = 5$  and  $u$  is 5 m/sec (a plausible value), then

$$\sigma/\sigma^{\text{LIGHT}} \cong (1 + v^2)^{-1/2}, \text{ and}$$

large droplets with  $v$ 's of nearly 10 m/sec are characterized by  $\sigma$ 's which are 1/10 of those for extremely small or light drops. The true  $\sigma$  is thus much smaller than one used in various CSL studies, including NUSSE2. Subcloud droplet concentrations are proportional to  $1/\sigma^3$ , but as we demonstrate below, deposition densities that are obtained by summing over all subclouds are roughly proportional to  $1/\sigma$ . These differences in  $\sigma$  lead to the discrepancies in deposition rates shown in Fig. 1.

### III. PRELIMINARY ANALYSIS

The purpose of this preliminary analysis is to explore the effect on the predicted deposition rate when the large fall velocities of C.W. droplets are included in the diffusion process. Therefore, we concentrate entirely on the dispersal and diffusion process to develop analytic formulas for purposes of comparison. Phenomena such as droplet evaporation, finite initial cloud dimensions, altitude variations in fall velocity and wind speed, vapor formation, surface evaporation, and dosage-time computations, which are included within the NUSSE2 model, are best analyzed via modifications to existing computer programs. It is not difficult to modify the CSL program to reflect a truer picture of the diffusion process. That portion of the NUSSE2 program dealing with surface evaporation may also require modification. As suggested earlier, an unrealistically large diffusion parameter could (because of other errors or omissions in the model) be obtained as a result of fitting NUSSE2 results to limited experimental data, but we have not yet analyzed the limited field data in sufficient detail to test this hypothesis.

#### THE GAUSSIAN PANCAKE MODEL

When a cloud is released, it contains a polydisperse mixture of droplets of different diameters and hence different vertical velocities. For convenience, we use a distribution of terminal fall velocities  $N(v)$ , where  $M_0 N(v)dv$  is the mass contained in the velocity interval,  $v, v + dv$ . A cloud thus consists of a continuum of subclouds, each with distinct fall velocity  $v$ . For concreteness, we choose a log-normal distribution for  $N(v)$ , with specified median velocity and standard deviation, but this is not essential. (Another approach, indicated below, is to employ a distribution for drop size and then relate terminal fall velocity to drop size.)

For a polydisperse Gaussian cloud or puff released at time  $t = 0$  at  $x = 0, y = 0, z = H$ , the concentration is



$$C(x, y, z, t) = \frac{M_0}{(2\pi)^{3/2}} \int_0^\infty \frac{N(v) dv}{\sigma^3} \exp \left[ - \left\{ \frac{(x - x_0(t))^2 + (z - z_0(t))^2 + y^2}{2\sigma^2} \right\} \right] \quad (4)$$

If the wind speed is constant,  $x_0(t) = ut$ , and if the specific fall velocity is  $v$ , then  $z_0(t) = H - vt$ . (Note that  $\sigma$  depends on both  $v$  and  $t$ .)

Let  $D(x, y)$  = total amount of liquid deposited per unit area at a point on the ground  $x, y$ . If  $vt > \sigma$ , or  $H > \sigma$ , then the surface concentration does not change appreciably as the cloud deposits on the surface. Physically it may then be shown that the total amount deposited on the element of surface area  $dx \cdot dy$  is equal to the liquid contained in a volume element whose base is  $dx \cdot dy$  and which extends from the bottom of the cloud to the top. Thus, each subcloud with a specific velocity  $v$  deposits liquid droplets as if it were a thin Gaussian pancake, with a deposition per unit area given by

$$\frac{M_0 \cdot N(v) dv}{2\pi\sigma^2} \exp \left\{ - \left[ \frac{(x - x_0(v))^2 + y^2}{2\sigma^2} \right] \right\}$$

where  $x_0(v)$  is the  $x$ -location of the centroid of the subcloud of fall velocity  $v$  as it impacts the ground. If the cloud is released at height  $H$ , time  $t = 0$ , and distance  $x = 0$ , then the subcloud impacts the ground at time  $t = H/v$ , and the  $x$  location of the this impact is  $x_0(v) = u \cdot H/v$ . We choose  $u$  as a characteristic wind speed, taken as  $1/H \int_0^H u dy$ , the average speed between 0 and  $H$ . This Gaussian pancake model is analogous to a tilted plume and to the model employed in Ref. 2.

As discussed earlier, the standard deviation,  $\sigma$ , is approximately related to  $v$  and  $t$  through

$$\sigma = \frac{2}{3} \cdot \frac{1^2 \cdot u^2 \cdot t}{\sqrt{v^2 + u^2/\beta^2}}$$

and if  $t$  is replaced by  $H/v$ , we obtain the specific  $\sigma(v)$  for the subcloud of velocity  $v$ .

$$\sigma(v) = \frac{2}{3} \frac{i^2 \cdot u^2 \cdot H/v}{\sqrt{v^2 + u^2/\beta^2}} \quad (5)$$

### SPATIAL DEPOSITION

The spatial deposition  $D(x,y)$  is obtained by integrating over the depositions from each subcloud or

$$D(x,y) = \frac{M_o}{2\pi} \int_0^\infty \frac{N(v) dv}{\sigma^2} \exp \left\{ -1/2 \left[ \frac{\left(x - \frac{Hu}{v}\right)^2 + y^2}{\sigma^2} \right] \right\} \quad (6)$$

If  $\sigma/H < 1$ , this may be approximated by<sup>1</sup>

$$D(\bar{x}, \bar{y}) \frac{H^2}{M_o} = \frac{u \cdot N\left(\frac{u}{\bar{x}}\right) \cdot \exp \left\{ -1/2 \left( \frac{\bar{y}}{\bar{\sigma}} \right)^2 \right\}}{\sqrt{2\pi} \cdot \bar{x}^2 \cdot \bar{\sigma}} \quad (7)$$

where  $D(\bar{x}, \bar{y}) \cdot H^2/M_o$  is the nondimensional deposition rate,  $\bar{x} = x/H$ ,  $\bar{y} = y/H$ ,  $\bar{\sigma} = \sigma/H$ , and

$$\bar{\sigma} = \frac{(2/3) \cdot i^2 \cdot \bar{x} \cdot \beta}{\sqrt{1 + \beta^2/\bar{x}^2}} \quad (8)$$

For a log-normal distribution in fall velocity,  $N(v)$  or  $N(u/\bar{x})$  may be written

<sup>1</sup>This formula is identical to the Porton model for the deposition of large droplets, except for our specification of  $\sigma$  in terms of fall velocity.

$$N(u/\bar{x}) = \frac{1}{\sqrt{2\pi} \cdot v \cdot (u/\bar{x})} \cdot \exp \left\{ - [\ln(u/\bar{x}) - \mu]^2 / 2v^2 \right\} \quad (9)$$

where  $\exp(\mu)$  is the median fall velocity and  $v$  is the standard deviation of  $\ln v$ .

To calculate the deposition rate, several parameters must be specified--two for the distribution of fall velocities  $\mu$  and  $v$ , and three for the atmospheric diffusion process,  $u$ ,  $\beta$  and  $i$ . Pasquill (as well as others) suggests that  $\beta$  (the ratio of the Eulerian to Lagrangian lengths scale) is between 1 and 10, with 5 being a widely quoted value. He also suggests that a turbulent intensity level  $i$  of 10 percent is reasonable for most atmospheric conditions. For preliminary comparisons, it is then acceptable to set  $\beta$  at 5 and  $i$  at .1. The current edition of Pasquill's classic treatise [1] suggests that  $\beta i$  may be roughly constant with a value close to .6. Presumably,  $\beta$  depends on meteorological conditions.

If  $v$  is terminal fall velocity and an empirical function  $\delta(v)$  is used to specify droplet diameter,  $\delta$ , in terms of fall velocity, then the mass distribution in velocity space may be rewritten in terms of droplet diameter or

$$N(v) = Q(\delta) \cdot \frac{d\delta}{dv} \quad (10)$$

when  $Q(\delta)$  is the mass distribution function in droplet diameter space. Given representations of  $Q(\delta)$  and  $\delta(v)$ , it is easy to determine the deposition. To illustrate, when  $Q(\delta)$  is log-normal in droplet diameter  $\delta$  with the standard deviation of  $\ln \delta$  equal to  $a$ , and the median droplet diameter equal to  $\exp(b)$ , the deposition relation now becomes

$$D(\bar{x}, \bar{y}) \cdot \frac{H^2}{M_0} = \frac{1}{2\pi} \cdot \frac{1}{\bar{x}} \cdot \frac{1}{\sigma a} \cdot \frac{d \ln \delta}{d \ln v} \exp \left\{ - \frac{1}{2} \left( \frac{\bar{y}}{\sigma} + \frac{(\ln(\delta) - b)^2}{a^2} \right) \right\} \quad (11)$$

Since the downwind motion of particles is assumed to be ballistic, the relation  $v = u/\bar{x}$  translates into a unique relation between  $\delta$  and  $\bar{x}$ , or  $\delta(v) = \delta(u/\bar{x})$ . Thus, the computation of  $D(\bar{x}, \bar{y})$  requires  $\delta(v)$ , the magnitudes of  $a$  and  $b$  to define the log-normal distribution, and Eq. (8) which defines  $\bar{\sigma}$ .

The illustrative results shown in Figs. 1 and 2 are based on Eq. (9) rather than Eq. (10). The curves are thus suitable for comparing relative deposition densities predicted by either neglecting or including fall velocity. Estimating the correct absolute magnitude of the deposition density would presumably require  $\delta(v)$  and the use of Eq. (11). As described below, Figs. 3 and 4 account for this in an ad hoc way.

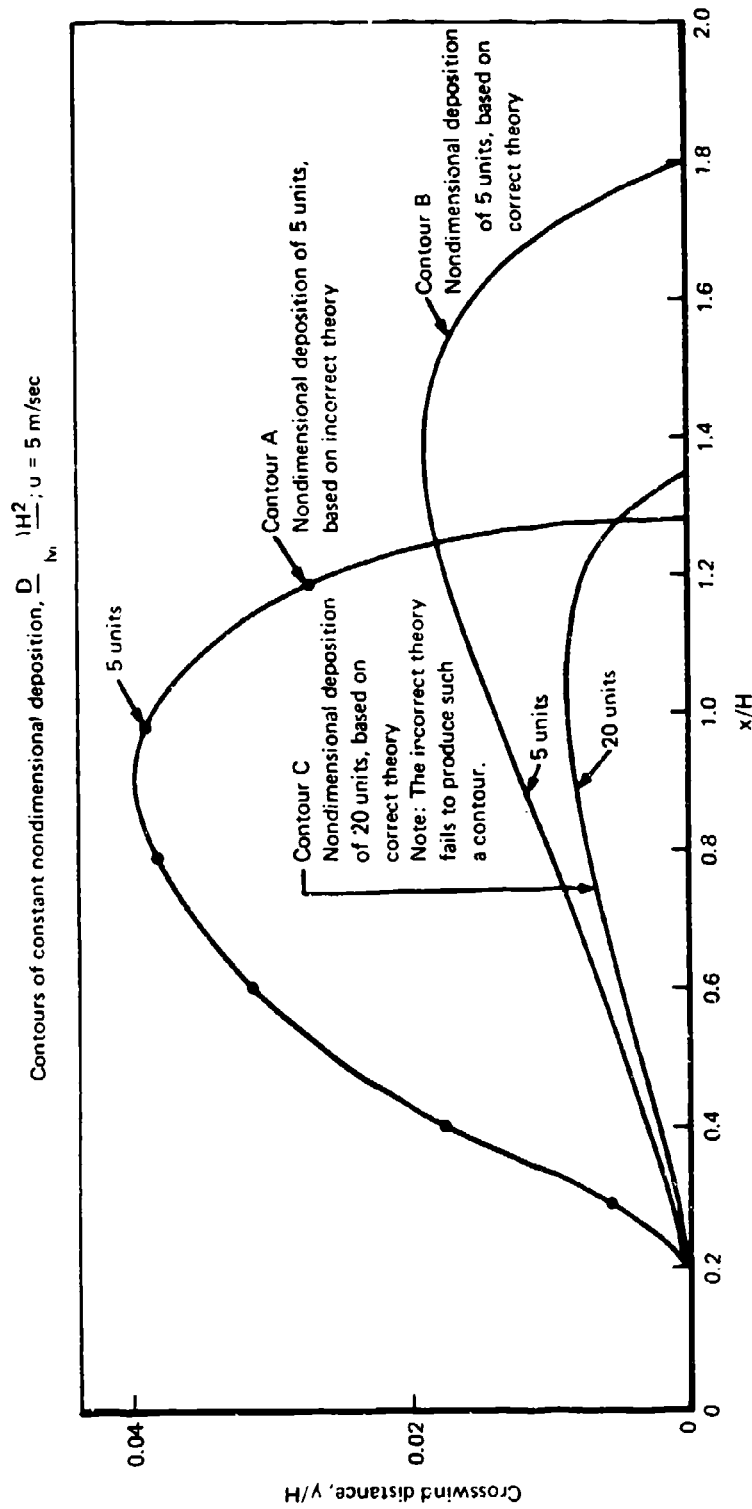


Fig. 2 — Contours of constant deposition

#### IV. RESULTS AND DISCUSSION

##### GENERAL

We have computed deposition patterns for a variety of wind speeds and distributions of fall velocity. Equations (7), (8), and (9) form the bases for Fig. 1, which indicates the deposition along the centerline ( $\bar{y} = 0$ ) when  $u = 5$  m/sec; the terminal velocity is characterized by a median of 5 m/sec with a standard deviation of 3.5 m/sec. As observed earlier, the discrepancy between curves A and B is large (over an order of magnitude). Roughly speaking, and when all other parameters are constant, the ratio of the "true" maximum deposition rate for heavy droplets to one computed using the inappropriate tracer particle diffusion value is (along the downwind direction  $\bar{y} = 0$ )

$$\sqrt{1 + \beta^2/x^2}$$

Thus, the true deposition rate is always greater than one computed using the "light" particle diffusion coefficient. Conversely, mass conservation requires that the true crosswind dimension of the contaminated region be correspondingly less than one estimated using the incorrect diffusion theory. This is demonstrated in Fig. 2, where several contours of constant spatial deposition density are shown. Contour A corresponds to a nondimensional deposition of five units computed using the incorrect NUSSE2 theory, and Contour B corresponds to the same rate of five units when computed using the more accurate theory. Note the difference in crosswise dimension. The "true" contamination region is narrower than the "incorrect" region.

An even more interesting and disconcerting discrepancy arises when we consider a contour corresponding to a deposition of 20 units. Such a contour (Contour C) may be found using the correct diffusion theory, but predicted deposition rates are always less than this value when the incorrect theory is used. Thus, the diffusion theory used in the CSL

computer program always underestimates the magnitude of liquid deposition, and yields low values for maximum exposure to toxic droplets. The practical importance of this is given by a hypothetical example: suppose the nondimensional value of 20 units corresponds to a biological threshold effect for a C.W. release of 500 kg at 10 km. The incorrect theory then results in a significant underestimate of hazard to personnel from liquid contamination. However, as we have speculated, although individual elements of C.W. defense models may be in serious error, the overall output of the models might correspond to test data obtained in limited field tests. This point deserves further study.

#### APPLICATION TO POWER LAW MODEL

For the sake of simplicity and theoretical consistency we have used a relation for  $\sigma$  based on Smith and Hay's theory for the diffusion of a falling monodisperse cloud or puff. (See appendix for further details.) Because there are few data about such clouds, and because turbulent diffusion theory is limited to idealized situations, some investigators prefer to use empirical diffusion coefficients and  $\sigma$ 's similar to those that describe the spreading of continuous plumes, such as smoke or particles issuing continuously from an elevated chimney. (As described in the appendix, the plume diffusion process differs from cloud or puff diffusion except at very long times.) In this spirit, NUSSE2 uses the plume formula  $\sigma(x) = A \cdot x^\alpha$ , where  $A$  and  $\alpha$  are empirical coefficients, presumably derived from the spreading of plumes of light tracer particles. In the spirit of Thomas' simplification of the Smith-Hay results (4), we suggest that a fall velocity correction for a puff would lead to

$$\sigma(x, v) = A \left[ \frac{x}{\sqrt{1 + \frac{v^2 \beta^2}{u^2}}} \right]^\alpha \quad (12)$$

This may be further simplified, when  $\sigma/H < 1$ , to

$$\sigma(x) = A \left[ \frac{x}{\sqrt{1 + \frac{\beta^2 H^2}{x^2}}} \right]^\alpha \quad (13)$$

where  $\beta$  is again assumed to be close to 5. This suggests a simple way to crudely correct for vertical velocity in the NUSSE2 results. A more rigorous correction procedure is only slightly more complicated.

Figures 3 and 4 demonstrate, in a practical context, the magnitude of the discrepancies between the correct and incorrect diffusion theories. Consider a release of 500 kg at 1.50 km. If the average wind speed is 7.1 m/sec, the downwind distribution of liquid deposition is maximum at about 1.1 km. However, the corrected maximum deposition rate is  $1.5 \times 10^4$  mg/mm<sup>2</sup>, whereas the uncorrected maximum is  $2.5 \times 10^3$ --roughly a factor of 5.6 (see Fig. 3). The crosswind effect is shown in Fig. 4--the crosswind distance of appreciable deposition shrinks from about 700 meters to 120 meters<sup>1</sup>--the deposition region shrinks and closely simulates a line source of material for subsequent evaporation.

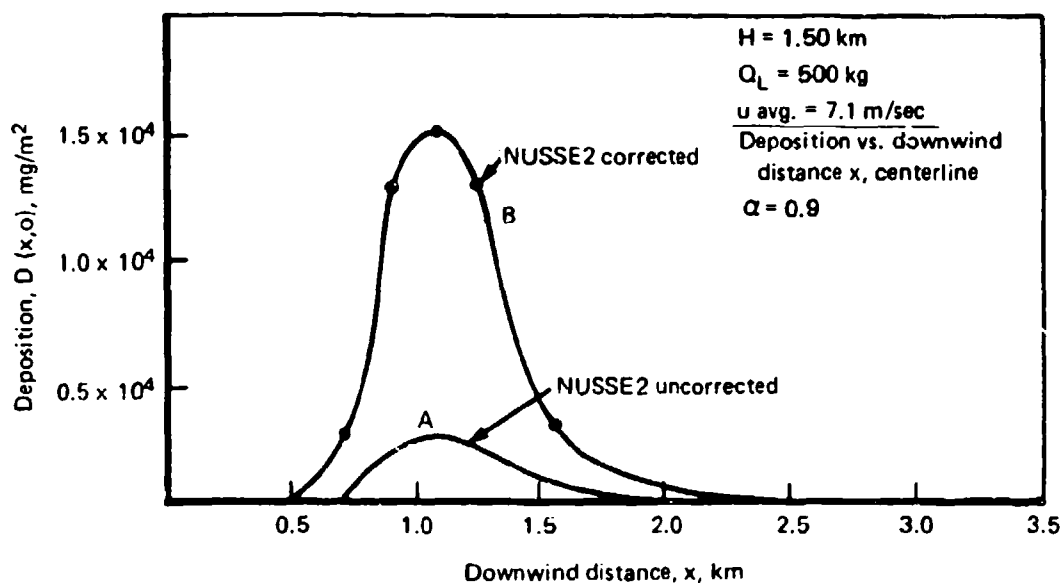


Fig. 3 -- Centerline deposition based on ad hoc correction

<sup>1</sup>From symmetry, total crosswind distance =  $2y$ .



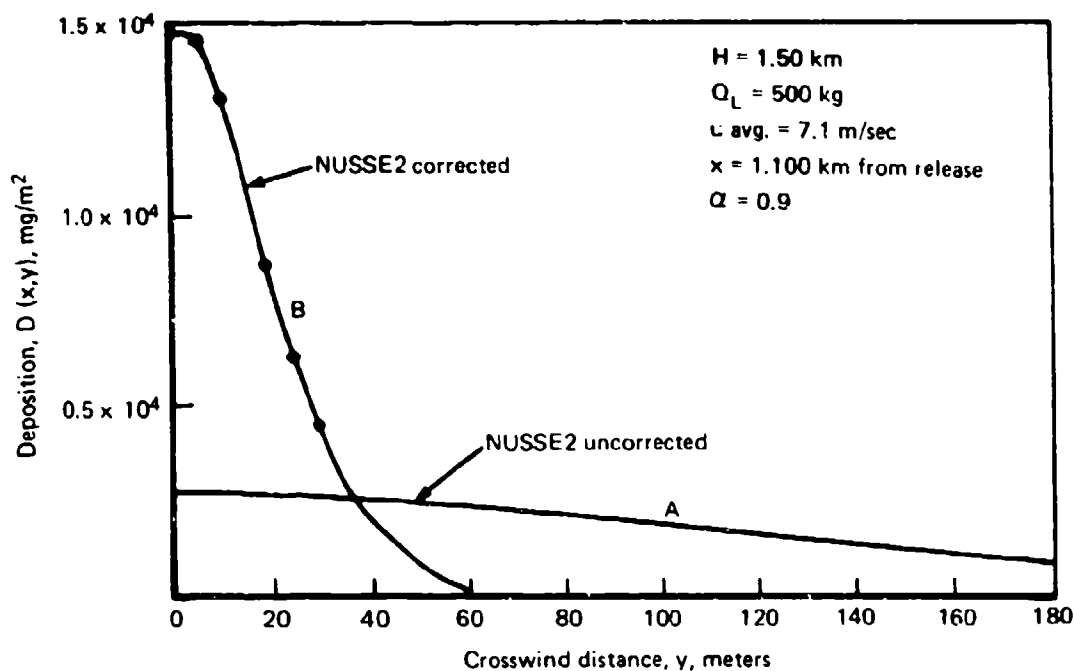


Fig. 4 - Crosswind deposition

As discussed above, we have not yet compared the results of "rigorous" theory to field test, and moreover have not found any relevant experimental data on falling clouds of heavy particles. However, both intuition and physics suggest that there would be an important reduction in diffusion.

If spuriously high diffusion parameters are used as adjustable constants ("fudge factors"), this would imply that other portions of the chemical deposition models are not described accurately, and that analysts must be wary when using NUSSE2 for cases that differ from the calibration cases derived from field data.

Several experiments have been performed in Canada that simulate the continuous (plume) diffusion of particles, using falling microspheres of various diameters. Superficially, these experiments suggest that crosswind diffusion is little influenced by fall velocity. But, as

indicated by Pasquill and Smith, plume diffusion is expected to be more weakly affected than puff diffusion, and the limited effects that are observed are quite consistent with theory. Furthermore, the microspheres used in the Canadian tests are considerably smaller than the probable droplet sizes that would result from the atmospheric dispersal of liquids carried by tactical missiles.

The discussion and analysis described in this Note are based on a decades old theory that has never been adequately tested against data. Because there is now a revival of interest in C.W. defense, and because the puff diffusion process for large droplets differs considerably from the diffusing of plumes of aerosols or atmospheric pollutants important in environmental studies (which have been studied intensively in recent years), we believe that now is the time to develop an appropriate experimental and analytic program. Such a program would include "research grade" experiments (using microspheres, or similar particles of known properties) and field tests, performed carefully and based on rheologically appropriate simulants.

A suitable program of test and analysis would provide:

- Comparison with experiment and field data.
- Confirmation of the diffusion theory discussed here for appropriate fall velocities using both alternative theories and Monte Carlo simulation. Dr. Steven R. Hanna has suggested (personal communication) that a Monte Carlo diffusion exercise could be conducted using velocity fields generated by a Large Eddy Simulation model, currently under study by an Army Research Office working group.
- Consideration of the impact of droplet inertia on atmospheric diffusion.
- Consideration of the liquid-water evaporation process on the ground, and the vapor exposure problem.
- Consideration of meteorological effects which are particularly important when releases within and above the atmospheric boundary layer are analyzed. (The CSL model employs a wind profile which is inappropriate for release above several hundred meters.) The influence of wind shear and the variation of wind direction with altitude should be included.

We remark, in closing, that such a program would both clarify the physics of droplet diffusion and narrow the uncertainty in models of C.W. defense.

## Appendix SURVEY OF PAST WORK ON THE DIFFUSION OF FALLING CLOUDS

### PUFF AND PLUME DIFFUSION

Studies of atmospheric diffusion distinguish between plume diffusion and puff diffusion. Plume diffusion refers to circumstances where material release and sampling times are long compared with travel time from the source, and puff diffusion refers to circumstances where material release and sampling times are short compared with travel time. Thus, an instantaneous source leads to puff diffusion and a continuous source to plume diffusion. The transport and diffusion of a cloud of material suddenly released in the atmosphere clearly corresponds to a puff diffusion process. The physics of puff diffusion is much less precisely characterized than that of plume diffusion. In the words of a recently published handbook [5], "We have a few theories for puff diffusion and a data set that is several orders of magnitude smaller than the data set for plume diffusion." Furthermore, the empirical literature on puff diffusion includes only limited data on the effect of atmospheric stability, and virtually no data on the dispersion of clouds of droplets or particles with appreciable fall velocity.

Analysts must then rely on a blend of theory, empiricism, and judgment. Despite the limited data, intuition suggests that the diffusion of clouds or clusters of heavy particles must be inhibited by the "trajectory crossing effect." This effect was first described by Soviet scientist M. I. Yudine [6]: "When falling, a heavy particle crosses trajectories of air particles so that it interacts consecutively with different air particles. As a result, the succession of velocities of a heavy particle does not coincide with individual changes of the velocity of an air particle." This leads to a reduction in dispersion.

Starting with Yudine, a number of investigators have analyzed plume diffusion of heavy particles, but only Smith [7] and Smith and Hay [3] have written about the puff diffusion counterpart. Puff diffusion is sensitive to a narrower range of turbulent eddy sizes than plume diffusion, being most influenced by eddies with sizes close to the puff

size. Similarity laws for puff diffusion have been developed by Batchelor, but such laws do not include the influence of fall velocity. For long times of flight of light particles, the Batchelor relation  $\sigma^2 = \epsilon t^3$ , where  $\epsilon$  is the eddy dissipation rate, has been recommended for practical use [5].

The puff theory for heavy particles first proposed by Smith, and later modified by Smith and Hay, starts with Batchelor's observation that puff diffusion depends on the relative motion of pairs of particles. Thus a two-particle Lagrangian covariance is required, whereas plume diffusion relies only on a single-particle Lagrangian covariance.

Smith and Hay assume homogeneous isotropic turbulence, invoke the Taylor transformation  $x = Ut$  to relate space and time covariances, and assume the validity of the Hay-Pasquill hypothesis to relate the Lagrangian time covariance to the Eulerian time covariance in terms of  $\beta$ , the ratio of the Lagrangian time scale to the Eulerian time scale. Ultimately the Smith-Hay equation for  $\sigma$ , in terms of the full spectrum of turbulence, becomes

$$\frac{d\sigma}{dt} = \frac{2}{3} \frac{\beta}{U} \int_0^\infty \int_0^{Ut/\beta} E(k) \frac{\sin ks}{ks} \cdot \frac{1 - e^{-\sigma^2 k^2}}{\sigma} ds dk \quad (A.1)$$

where  $E(k)$  is the energy density.

If  $Ut/\beta$  is large enough, then further analysis (after assuming an exponential correlogram) leads to

$$\frac{d\sigma'}{dX'} = 2\beta i^2 \int_0^\infty \frac{n^2}{(1+n^2)^3} \frac{1 - e^{-(\sigma')^2 \cdot n^2}}{n\sigma'} dn \quad (A.2)$$

where  $\sigma' = \sigma/\ell$ ,  $X' = X/\ell$ ,  $i$  is turbulence intensity and  $\ell$  is the Eulerian length scale.

If  $Ut/\beta$  is not large enough to be accurately replaced by infinity, the covariance approach leads to

$$\frac{d\sigma'}{dX'} = \beta l^2 E\left(\sigma', \frac{X'}{\beta}\right) \quad (A.3)$$

where  $E$  is a function graphed on p. 92 of Ref. 3. Two further approximations for  $\sigma$  are also developed:

1. If the initial value of  $\sigma'$  is sufficiently small, then  $\sigma'(0)$  may be set equal to zero during the integration of Eq. (A.2), and a universal curve is obtained for initially small puffs (or clusters). This curve portrays a relation  $\frac{\sigma}{l} = F\left(\frac{\beta l^2 X}{l}\right)$ , where  $X$  is measured from an arbitrary origin.
2. A further simplification results after noting that  $E$  in Eq. (A.3) may be replaced with (reasonable accuracy) by its maximum value, leading to Thomas' approximation [4].

$$\frac{d\sigma}{dX} = \frac{2}{3} \beta l^2 \quad (A.4)$$

These  $\sigma$ 's correspond to  $\sigma^{\text{LIGHT}}$ , and refer to the diffusion of light particles.

## DIFFUSION OF HEAVY PARTICLES

The results discussed above were originally derived for clusters of light particles. However, they may be extended to clusters of particles that have sufficient mass to acquire appreciable terminal velocity, but are still small enough to respond to the local turbulent velocity fields.

Under these conditions, where particle inertia may be neglected, an approximate equivalence principle is described by Pasquill and Smith [1], who note that a falling particle moves horizontally with the fluid and experiences Lagrangian variations equivalent to passing through the space spectrum with velocity  $U/\beta$ , and simultaneously falls through the space spectrum with velocity  $v$ . The result, they suggest, is equal to moving through the space spectrum with velocity  $(v^2 + U^2/\beta^2)^{1/2}$ .

Frequencies at a fixed point then appear to be increased in the proportion  $(v^2 + U^2/\beta^2)^{1/2}/U$ , and the integral time scale is correspondingly decreased.

When  $v$  is nonzero, the parameter  $\beta$  in Eqs. (A.1), (A.2), (A.3), and (A.4) may be replaced by  $\beta' = \{(v/U)^2 + 1/\beta^2\}^{-1/2}$ . For example, when  $\sigma(0)$  is small,  $\sigma/\ell$  may be represented by

$$\frac{\sigma}{\ell} = F \left[ \left\{ \frac{v}{U} + \frac{1}{\beta^2} \right\}^{-1/2} \cdot i^2 \cdot \frac{x}{\ell} \right] \quad (A.5)$$

and Eq. (A.4) now becomes

$$\sigma = \frac{2}{3} \cdot \left[ \left( \frac{v}{U} \right)^2 + \frac{1}{\beta^2} \right]^{-1/2} \cdot i^2 \cdot x. \quad (A.6)$$

Furthermore, the validity of replacing  $\beta$  by  $\beta'$  is independent of the particular form of the correlation function. Thus, Sawford's recent study of relative diffusion [8], using alternative representations of the two-particle Lagrangian correlation function, may easily be modified to include the influence of fall velocity.

The heavy particle effect is greater for puff diffusion than for plume diffusion. For example, Csanady's plume theory [9] leads to

$$\frac{\sigma_v}{\sigma_{\text{LIGHT}}} = \left( 1 + \frac{\beta^2 v^2}{U^2} \right)^{-1/4} \quad (A.7)$$

and the Smith-Hay theory (and Thomas' approximation) for puff diffusion leads to

$$\frac{\sigma_v}{\sigma_{\text{LIGHT}}} = \left( 1 + \frac{\beta^2 v^2}{U^2} \right)^{-1/2} \quad (A.8)$$

For droplets which fall with terminal velocities as large as 10 m/sec, we see that the standard deviation is 30 percent of the light particle value for a plume, and 10 percent of the light particle value for a puff, when windspeed is 5 m/sec and  $\beta \approx 5$ .

The parameter  $\beta$  is not very well characterized--some analysts now believe that  $\beta$  is a constant of unit order (say .6), and others have tried to infer a dependence of  $\beta$  on stability.

As far as experimental verification is concerned, there does not appear to be any field data for falling clusters that may be used to evaluate or modify the Smith-Hay data. Thomas alluded to such data in his 1964 note, but we have not yet found any in the open literature.

Because the characteristic dimension  $\sigma$  for falling clusters is much less than for clusters of light or tracer particles, we speculate that interactions between wind shear, either horizontal or vertical, and diffusion are less important than for light particles.

Particle inertia, and its effect on the diffusion of falling droplet clouds, is a virtually unexplored area. The Smith-Hay theory is based on the assumption that inertial effects are not important except to define the fall velocity of the particle. Certainly, we expect that inertia is negligible for droplets in the submillimeter range (based on plume diffusion results).

We are less certain about larger droplets in the 1 to 10 mm size range. Analysis by Reeks [10], and Nir and Pismen [11], indicate that particle inertia has little effect on plume (absolute) diffusion when the turbulent velocity is smaller than the velocity of fall, and that Csanady's formula, (A.5), is valid over a broad range in particle inertia.

Thus, although particle inertia may inhibit the large effect of fall velocity in reducing diffusion, the magnitude appears to be small for plume diffusion. Until the influence of inertia on the puff diffusion process has been analyzed properly, we therefore believe that its neglect may be justified.

In summary, then, there is but one theory, and no "research grade" data to define the diffusion of clusters of falling particles. The theory is physically plausible and indicates an important diminution of



dispersion for heavy particles. Verification and extensions of the theory remain to be pursued. Clearly, those who are interested in accurately modelling the diffusion of toxic droplet clouds should now use formulas like (A.5) and (A.6), or the Pasquill-Smith  $\beta'$  described above, to modify their representations for  $\sigma$ , since, as shown in the body of the Note, the heavy-particle effect can lead to an order of magnitude difference in deposition.

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